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Research progress of microstructure control for aluminium solidification process

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Aluminum has been the secondly main metallic material in the world, whose mechanical properties are very important for industrial applications. The sizes and shapes of grains are important in determining the performance in structural applications. How to control the microstructure during solidification process has been a research focus. This paper gives an overview on the recent progress in microstructure control for aluminium alloys solidification process, and introduces the different methods to control the microstructure in detail. The mechanisms of microstructure control for different methods are also discussed. Finally, a brief prospect on future work is presented.

aluminium alloy, microstructure control, solidification, grain refinement

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Steel is the most widely used in structural and functional applications, and high performance steels have been a new research focus in recent years [1–10]. Aluminum is the richest metallic element in the earth, which has been the secondly main metallic material in the world. Fracture properties of materials are very important for industrial applications, and different mathematical models have been developed to simulate the fracture of materials [11–14]. For aluminium alloys, the sizes and shapes of the grains can be important in determining the performance in structural applications. Fine grain size is beneficial to aluminium alloys for ambient temperature structural use, because it simultaneously gives higher strength and greater toughness. Additionally, grain refinement can decrease the size of defects and increase the resistance to hot tearing, especially for producing large, superhigh aluminium alloy ingots [15]. On the other hand, single crystal with almost no grain boundary and nanomaterials are important in determining the performance of functional applications [16–18]. Zhang et al. [19] simulated the microstructure evolution during the directional solidification process. Aluminum foam with low density is attractive as cushion material, and which can be used for mitigating shocks, absorbing impact energies, etc. [20]. Additionally, amorphous alloys are the recent developed metallic materials, and exhibit many excellent properties [21].

As mentioned above, metal materials are used widely in human society, and plenty of research has been done in material science recently [22]. The sizes and shapes of the grains are crucial for the mechanical properties, and they are determined by the solidification process. Solidification process is a phase transition, in which a liquid turns into a solid when its temperature is lower than its freezing point. It occurs in a wide range of industrial processes, including conventional casting and single crystal growth. In order to reveal the solidification process intuitively, transparent model materials can be used to study the solidification process [23]. The melt flow during solidification process can be seen as the fluid flows with different viscosities under different temperatures. The porous media are usually used to simulate the melt flow during the solidification process. Re-

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searches about viscosity, convection and solution of equation have been carried out, and different meshing technologies, principles and models have been developed for finite element analysis [24–33].

In some practical cases, the final grain size and microstructure are the result of the solid state process. The recrystallization following mechanical deformation and deformation temperature are important for the plastic deformation process [34–36]. In this paper, we mainly discuss the grain control of as-solidified structure during the aluminium alloys solidification process. For solidification structure control, there are many methods such as adding alloy elements, adding grain refiner, applying the physical fields, etc.

1 Structure control by adding alloy elements

Many experimental results show that adding some alloy element into the aluminium alloy can affect the structure and improve the mechanical properties. There are a wide variety of alloy elements, and their effects and mechanisms are different. How to choose the kind of elements and how to control the content of elements to obtain optimum alloying effect have been investigated widely. The rare earth elements and transition elements are usually chosen to alloy the aluminium alloy. The second phase particles formed by the rare earth element or transition element can be heterogeneous nucleation. The solidification structure of aluminium alloy can be refined, the growth of grain can be restrained during the recrystallization process, and the aluminium alloy with good superplasticity can be obtained [37].

The addition of rare earths in aluminium alloys can refine the grains, purify the melt and reduce the gas and inclusion contents. So, the comprehensive properties of aluminium alloys are improved [38]. The existing researches show that the alloying effect of Sc is the most obvious among all the alloying rare earth elements. When the Sc element is added into the Al–Cu–Mg–Ag alloy, approximately 70% of the Sc added into aluminium alloy are fixed in the supersaturated solid solution. Two thirds of Sc in the supersaturated solid solution are consumed by the formation of a dispersion of nanoscale Al_3Sc particles within the Al matrix during the homogenisation annealing process. A third of the Sc are consumed as the formation of the W-phase because of Sc diffusion into the primary θ-phase, which leads to its transformation into the W-phase [39]. However, the Sc element is very expensive, so the aluminium alloys containing rare earth element Sc are too expensive to be widely used in industrial areas. Nie and his group [40] found that rare earth element Er is a cheap and effective alloying element for aluminium alloy. The nano-scale Al_3Er particles can form in aluminium alloys containing Er, and the composite phase $Al_3(Zr_xEr_{1-x})$ can form due to the interaction of Er and Zr in the liquid metal of a high temperature. The Al_3Er and $Al_3(Zr_xEr_{1-x})$ particles are thermally stable, thus the addition of rare earth element Er can modify the microstructure and improve the mechanical properties. Additionally, the Al_3Er and $Al_3(Zr_xEr_{1-x})$ particles can also hinder the recrystallization and improve its thermal stability [40].

The transition elements (V, Zr, etc.) can be added into the liquid metal to refine the grains and improve the fine-grained superplasticity. The research results show that the trace element vanadium added into 5083 alloy can refine the cast structure and fibrous structure of the rolling sheet, restrain the grain growth during recrystallization, and improve the superplasticity of 5083 aluminium alloy. The size of recrystallized grains of the sheets was reduced from 100 to 20 μm as shown in Figure 1, where the high temperature superplastic performance of 5083 sheet was improved and the elongation percentage of 5083 aluminium alloy sheet in 510°C was improved from 208% to 254% after the addition of trace element vanadium [41]. The transition element Zr is also usually added into high strength aluminium alloys, and the Al_3Zr particles can precipitate in grain boundary areas during the homogenization treatment process. The presence of Al_3Zr particles can promote continuous dynamic recrystallization during the hot extrusion process, and a fine-grained structure can be obtained. The Al_3Zr particles can pin the grain boundary migration. A large flow stress is required for the grain boundary mobility to increase as the amount of Zr addition increases [42]. The Al_3Zr particles precipitate in the grain boundary areas, which can lead to the hot-rolling textures. The Al_3Zr particles which are coherent with aluminium matrix can inhibit the movement of dislocation induced by the plastic deformation. So, the Al_3Zr particles can decrease the deformation textures and prevent recrystallization, which can increase the material hardness and extend the existence time of peak hardness [43].

The rare earth element Sc and transition element Zr can also be combined to add into the aluminium alloy [44]. Lidia and his groups [45] added the Sc and Zr elements into the Al-12Zn-3Mg-1.5Cu alloy. The experimental results showed that a significant grain refinement was observed as the presence of fine primary $Al_3(Zr, Sc)$ particles when 0.2%Zr and 0.3%Sc were added into the aluminium alloy.

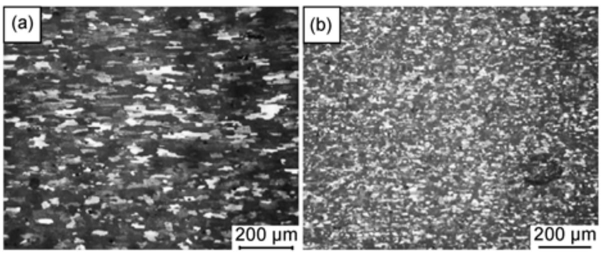

Figure 1 Microstructures of undeformed region. (a) 5083 alloy, (b) 5083 alloy with trace element vanadium.

The additions of Sc and Zr can result in the increase of the microhardness. The heat treatment at 400–460°C can increase the microhardness due to the dissolution of alloying elements into the Al solid solution and the precipitation of $Al_3(Zr, Sc)$ particles in the Zr and Sc contained ribbons [45].

2 Structure control by grain refiners

The earliest grain refiner master alloy used in aluminium alloys is the Al-Ti master alloy. On the basis of Al-Ti master alloy, Al-Ti-B master alloy was developed. The effect of grain refinement is improved greatly. However, the density of second phase particles ($TiAl_3$ and TiB_2) in the liquid metal is greater than that of the liquid aluminium alloy. The $TiAl_3$ and TiB_2 particles can agglomerate into larger particles during the standing process, and the effect of grain refinement can be invalid six hours after adding them into the liquid metal. In order to avoid the agglomeration of TiB_2 during the standing process, Al-Ti-C, Al-Ti-C-B, Al-Ti-C-RE and Al-Ti-B-RE master alloys were developed [46–48]. Doheim et al. [49] evaluated the Al-Ti-C master alloys as grain refiner for aluminium alloys. The second phase particles (TiC) can exist steady and not agglomerate into larger particles. The addition of rare earth element can improve the morphology and distribution of the TiB_2 and $TiAl_3$, refine the size of $TiAl_3$ particles, increase the number of heterogeneous nuclei and improve the refinement effect of Al-Ti-B [50]. However, the elements such as Cr, Zr, Mn, etc. can interact with the Ti element. The grain refining test results showed that the presence of Zr in commercial aluminium melts can reduce the effectiveness of Al-5Ti-B master alloy [51].

Quantitative analyses confirmed that the reduction in growth restriction is not the sole reason for poisoning action in the melt. The Zr element can substitute for titanium in the aluminide and affect the nucleation stage of grain refinement. The existence of Al_3Ti on TiB_2 particles is the key to the nucleation potency [52]. Zhang and his group [43] found that the Al_3Zr particles can combine with Al_3Ti particles both in Al-Ti-Zr-C and Al-Ti-Zr-B alloys easily. The potency of Al_3Ti acting as nucleation particles and grain refiners is impaired and results in the poisoning action. While the Al_3Zr particles seem not to integrate with TiC and TiB_2 particles though the agglomeration of TiB_2 particles exists. The calculation results by using the edge-to-edge matching model indicated that Al_3Zr has a good crystallographic matching with Al_3Ti and α-Ti particles. The matching with Al_3Ti and α-Ti particles is better than that with TiC and TiB_2 particles. It means that the quantities of the orientation relationships between the matrix and precipitate can be used as a grain refiner for the crystallography theoretical consultation [53].

3 Structure control by applying the physical fields

The solidification process of liquid metal is a complex interplay containing many physical effects, and the liquid/solid interface is a free boundary. The latent heat is liberated and conducted away from the interface through the solid and liquid during the solidification process. The pressure on the liquid/solid interface, solidification rate and liquid/solid ratio are the main factors to affect the grain growth, solidification process and structure [54–58]. The melt flow, pressure and temperature distribution can be changed by the mechanical stirring [59] electromagnetic field [60] and ultrasonic wave [61] caused by applying the physical fields, and the solidification structure can be modified.

Mechanical stirring is strong, and always be used for liquid metal treatment. Fan and his group [59] found that the oxide films entrained within melts can be dispersed uniformly in the melt by an intensive stirring and shearing. The dispersed oxide films can act as nucleation centres, so the grain size can be reduced without any addition of grain refiner [59]. Based on the recent melt conditioning by advanced shear technology (MCAST), Zuo et al. [62] carried out an intensive melt shearing on 7032 aluminium alloy. The possibility to transform oxide films from potential defects into potent grain refiners was evaluated by an intensive melt shearing. The experimental results showed that the intensive melt shearing can refine the grains greatly. Additionally, the intensive melt shearing can reduce the size of porosity. The density index can be reduced and the average size of porosity in the samples solidified under partial vacuum is also reduced. For mechanical stirring, the stirring paddle contacts the liquid metal directly, and the stirring paddle could interact with the melt when the stirring paddle is rotating with a high speed and the liquid metal could be polluted.

Electromagnetic stirring is one of the key technologies which can be used to control the solidified structure without contact with liquid metal. The early development of electromagnetic casting (EMC) was described by Getselev [63]. A high frequency electromagnetic field was supplied in order to produce an electromagnetic pressure which was used to balance the static pressure. As there was no contact between the mold and the melt, ingots with good surface quality were obtained and the structure and segregation could be changed by using this technology. The casting, refining and electromagnetic process (CREM) [64] can both refine microstructure and improve surface quality of ingots. The coil surrounded the ingot mold and an alternating current of standard industrial frequency (50 or 60 Hz) was supplied. Under the effect of the periodic current, the inductor generates an alternating magnetic field and the melt can be inductively stirred. The constrained effect of electromagnetic forces reduces the contact height between the melt and mold, but the dome height which influences the thickness of the segregation is hard to control.

Based on the CREM process, a lower frequency electromagnetic field was applied in the direct chill casting process by Cui and his coworkers [65] to control the microstructure of the aluminium alloy ingot by the higher penetrability of lower frequency electromagnetic field in metallic melt. To improve the quality of 7050 aluminium alloy ingots, a low frequency electromagnetic field was applied during the conventional hot-top casting process. The experimental results showed that when the lower frequency electromagnetic field is turned off during the hot-top casting process, cold folding appears, and the as-cast structure becomes very coarse as shown in Figure 2. Moreover, the thickness of the shell zone is much thinner during the low frequency electromagnetic hot-top casting process than during the conventional hot-top casting process [66].

When a low frequency electromagnetic field is applied during the casting process, the alternating current generates a time varying magnetic field in the melt, which gives rise to an induced current in the melt. Therefore, the melt is subjected to electromagnetic body forces caused by the interaction of the induced current and the magnetic field. Another characteristic of the electromagnetic field is the presence of a fringe effect consisting of a pronounced inclination of the magnetic field lines toward the axis of symmetry of the ingot. Therefore, the Lorentz force consists of two parts expressed as follows:

$$F = -\frac{1}{2\mu}\nabla(B)^2 + \frac{1}{\mu}(B\cdot\nabla)B, \tag{1}$$

where F, B and μ are Lorentz force density vector, magnetic flux density vector, and permeability, respectively. The first part of eq. (1) is a potential force balanced by the static pressure of the melt (Fr), the second part is a rotational component which results in a forced convection in the melt (Fz). The natural convection near the mold wall can be changed into a forced convection by this rotational force (Fz). The flow of molten metal under the overhang is intensified, the liquidity of the melt near the meniscus becomes good, and the "dead zone" with poor liquidity is minimized or even disappears. So, the depth of cold fold is reduced as shown in Figure 2.

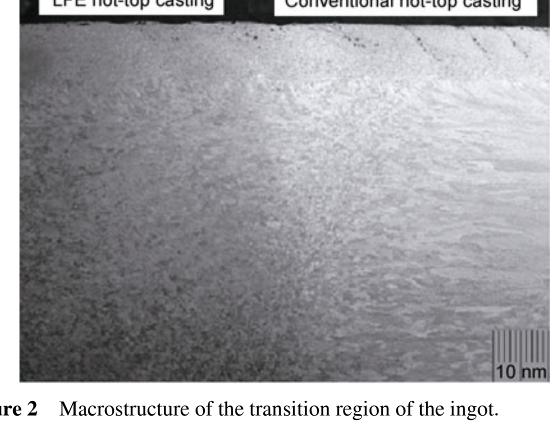

Figure 2 Macrostructure of the transition region of the ingot.

4 Conclusions

The properties of aluminium alloys are crucial for the industrial application. The sizes and shapes of grains are important in determining the performance of aluminium alloys. The modification of microstructure during solidification process has been a research focus. Adding alloy elements, using grain refiners and applying physical fields are the main methods for controlling the solidification structure. In this paper, we presented an overview of the methods for microstructure control. Several alloy elements such as Sc, Er, Zr, V, etc. used for microstructure control were listed and the main grain refining mechanism of alloy elements was illustrated. Some grain refiners and the poisoning of the grain refiner action after addition of 0.05 wt% Zr were introduced. Furthermore, the mechanical field can be used to condition the melt prior to solidification, and electromagnetic field can be used to stir the melt during the solidification process. This review demonstrated that the microstructure and properties of aluminium alloys can be controlled by different methods. However, there are still some technical problems to deal with. New alloy elements, grain refiners and physical fields for microstructure control can be developed by future solidification researches.

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